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(translation)

AMENDMENT

(Amended under PCT Article 34)

To Mr. Mituji UEMAE, Examiner of the Japan Patent Office

1. Identification of the International Application:

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4. Subject of Amendment:

Specification and Claims

5. Content of Amendment:

As in the attached document

6. Item of Attached Document(s):

- (1) Pages 4 to 6, 28, 32, 34 to 35, 41 to 43, 51 to 52, 56 to 57 and 62 of Specification (Page numbers 5 to 8, 36, 37, 42, 44, 45, 45/1, 47, 47/1, 56, 56/1, 58, 58/1, 68, 69, 69/1, 76, 78, 78/1 and 84 as in the attached document)
- (2) Pages 63 to 66 and 66/1 of Claims (Page numbers 85 to 91 and 91/1 as in the attached document)

## Content of amendment

(1) All the description on page 4, paragraph [0011] of the specification (page 5, line 16 to page 6, line 13 of the English specification) is amended as follows:

“Accordingly, the present invention provides a dye-sensitized solar cell module comprising: first photoelectric conversion elements each comprising a transparent conductive layer, a porous photoelectric conversion layer adsorbing a dye, an electrolytic layer, a catalyst layer, and a conductive layer laminated in this order on a transparent substrate; second photoelectric conversion elements each comprising a transparent conductive layer, a catalyst layer, an electrolytic layer, a porous photoelectric conversion layer adsorbing a dye, and a conductive layer laminated in this order on the transparent substrate; and a supporting substrate formed on the respective conductive layers of the first and second photoelectric conversion elements, wherein one or more first photoelectric conversion elements and one or more second photoelectric conversion elements are alternately arranged in parallel between the transparent substrate and the supporting substrate, and the neighboring first photoelectric conversion elements and second photoelectric conversion elements are electrically connected in series, and wherein the first photoelectric conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the materials for the respective layers composing the photoelectric conversion elements; the particle diameters on the case that the materials of the respective layers are granular; the thickness and width of the respective layers; the form of the respective layers; and the dye.”

(2) All the description on page 5, paragraph [0012] of the specification (page 6, lines 15 to 27 of the English specification) is amended as follows:

“In the dye-sensitized solar cell module of the present invention, the first photoelectric conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the materials for the respective layers composing the photoelectric conversion elements; the particle diameters on the case that the materials of the respective layers are granular; the thickness and width of the respective layers; the form of the respective layers; and the dye, so that the properties such as the photoelectric conversion efficiency and the electric current can be improved as compared with a conventional technique.”

(3) All the description on page 6, paragraph [0015] of the specification (page 7, line 19 to page 8, line 21 of the English specification) is amended as follows:

“According to an embodiment of the present invention, there is provided a dye-sensitized solar cell (hereafter, sometimes referred to simply as a solar cell) comprising: first photoelectric conversion elements each comprising a transparent conductive layer, a porous photoelectric conversion layer adsorbing a dye, an electrolytic layer, a catalyst layer, and a conductive layer laminated in this order on a transparent substrate; second photoelectric conversion elements each comprising a transparent conductive layer, a catalyst layer, an electrolytic layer, a porous photoelectric conversion layer adsorbing a dye, and a conductive layer laminated in this order on the transparent substrate; and a supporting substrate formed on the respective conductive layers of the first and second photoelectric conversion elements, wherein one or more first photoelectric conversion elements and one or more second photoelectric conversion elements are alternately arranged in parallel between the transparent substrate and the supporting substrate, and the neighboring first photoelectric conversion elements and second photoelectric conversion elements are electrically connected in series, and wherein the first photoelectric

conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the materials for the respective layers composing the photoelectric conversion elements; the particle diameters on the case that the materials of the respective layers are granular; the thickness and width of the respective layers; the form of the respective layers; and the dye.”

(4) On page 28, paragraph [0067], lines 5 to 6 of the specification (page 36, lines 24 to 29 of the English specification), the phrase “light-irradiated face” is amended to --light receiving face--, and the phrase “conductive layer side” is amended to --porous photoelectric conversion layer side--.

(5) On page 28, paragraph [0068], Table 6 of the specification, the phrase “conductive layer side” is amended to --porous semiconductor layer side--.

(6) On page 32, paragraph [0078], lines 8 to 9 of the specification (page 42, lines 25 to 28 of the English specification), the phrase “light-irradiated face” is amended to --light receiving face--.

(7) On page 34, lines 1 to 2 of the specification (page 44, line 24 of the English specification), the phrase “single dye-sensitized solar cell” is amended to --cell unit--.

(8) On page 34, paragraph [0083], line 6 of the specification (page 45, lines 5 to 6 of the English specification), the phrase “the light receiving face and set in the catalyst layer side” is amended to --the light receiving face is set in the catalyst layer side--.

(9) On page 35, paragraph [0086], line 8 of the specification (page 47, line 19 of the English specification), the word “cetonitrile” is amended to

--acetonitrile--.

(10) On page 41, paragraph [0103], line 5 of the specification (page 56, line 9 of the English specification), the word “cetonitrile” is amended to --acetonitrile--.

(11) On page 42, paragraph [0105], lines 2 and 5 of the specification (page 56, lines 23 and 28 of the English specification), the phrase “Comparative Example 6” is amended to --Comparative Example 5--.

(12) On page 43, paragraph [0108], lines 2 and 5 of the specification (page 58, lines 10 and 15 of the English specification), the phrase “Comparative Example 7” is amended to --Comparative Example 6--.

(13) On page 51 of the specification, the phrase “the example” on line 7 (page 68, line 12 of the English specification) is amended to --Example 21--, and the phrase “the dye 7” on lines 7 to 8 (page 68, line 12 of the English specification) is amended to --the black dye defined by the above-mentioned formula (II)--.

(14) On pages 51 to 52, paragraph [0122] of the specification: the phrase “Comparative Example 8” on lines 2 and 6 (page 69, lines 7 and 15 of the English specification) is amended to --Comparative Example 7--; the phrase “Example 17” on line 5 (page 69, line 13 of the English specification) is amended to --Example 21--; the phrase “Comparative Example 6” on line 5 (page 69, line 14 of the English specification) is amended to --Comparative Example 7--; and the phrase “coult be produced” on lines 9 and 10 (page 69, line 20 of the English specification) is amended to --could be produced--.

(15) On page 56, paragraph [0132], line 5 of the specification (page 76, line 11 of the English specification), the phrase “Comparative Example

9” is amended to --Comparative Example 8--.

(16) On page 56, paragraph [0133], line 1 of the specification (page 76, line 25 of the English specification), the phrase “Comparative Example 9” is amended to --Comparative Example 8--.

(17) On page 57, paragraph [0136], line 4 of the specification (page 78, line 2 of the English specification), the phrase “Example 18” is amended to --Example 27--.

(18) On page 62, paragraph [0151], line 4 of the specification (page 84, line 7 of the English specification), the phrase “Comparative Example 10” is amended to --Comparative Example 9--.

(19) All the description in claim 1 on page 63 is amended as follows:

“A dye-sensitized solar cell module comprising:

first photoelectric conversion elements each comprising a transparent conductive layer, a porous photoelectric conversion layer adsorbing a dye, an electrolytic layer, a catalyst layer, and a conductive layer laminated in this order on a transparent substrate;

second photoelectric conversion elements each comprising a transparent conductive layer, a catalyst layer, an electrolytic layer, a porous photoelectric conversion layer adsorbing a dye, and a conductive layer laminated in this order on the transparent substrate; and

a supporting substrate formed on the respective conductive layers of the first and second photoelectric conversion elements,

wherein one or more first photoelectric conversion elements and one or more second photoelectric conversion elements are alternately arranged in parallel between the transparent substrate and the supporting substrate, and the neighboring first photoelectric conversion elements and second photoelectric conversion elements are electrically connected in series, and



wherein the first photoelectric conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the materials for the respective layers composing the photoelectric conversion elements; the particle diameters on the case that the materials of the respective layers are granular; the thickness and width of the respective layers; the form of the respective layers; and the dye.”

(20) Claim 2 on page 63 is cancelled.

(21) All the description in claim 3 on page 63 is amended as follows:

“The dye-sensitized solar cell module according to claim 1, wherein a short circuit current of the second photoelectric conversion elements in the case where a light receiving face thereof is set in the porous photoelectric conversion layer side opposite the catalyst layer side is greater than a short circuit current of the first photoelectric conversion elements in the case where a light receiving face thereof is set in the porous photoelectric conversion layer side opposite the catalyst layer side.”

(22) All the description in claim 4 on page 63 is amended as follows:

“The dye-sensitized solar cell module according to claim 1 or 3, wherein the first photoelectric conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the electrolytic layers; the thickness of the porous photoelectric conversion layers; the width of the porous photoelectric conversion layers; the average particle diameter of the semiconductor particles composing the porous photoelectric conversion layers; and the form of the catalyst layers.”

(23) All the description in claim 7 on page 64 is amended as follows:

“The dye-sensitized solar cell module according to any one of



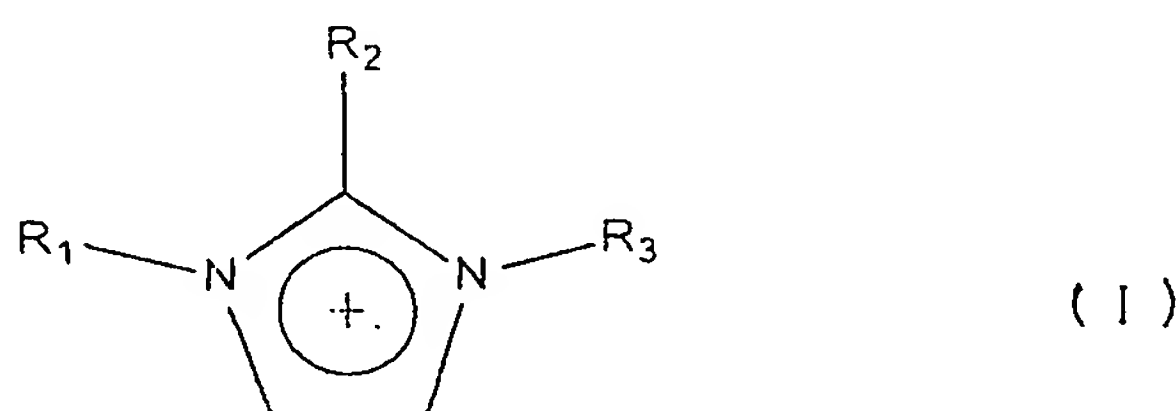
claims 1 and 3 to 6, wherein the first photoelectric conversion elements and the second photoelectric conversion elements respectively contain imidazoles or imidazolium salts in their electrolytic layers, the imidazole or imidazolium salt contained in each of the electrolytic layers of the first photoelectric conversion elements being different from that contained in each of the electrolytic layers of the second photoelectric conversion elements.”

(24) All the description in claim 8 on page 64 is amended as follows:

“The dye-sensitized solar cell module according to any one of claims 1 and 3 to 7, wherein the imidazoles or imidazolium salts contained in the respective electrolytic layers of the first photoelectric conversion elements and the second photoelectric conversion elements differ in concentration.”

(25) All the description in claim 9 on page 64 is amended as follows:

“The dye-sensitized solar cell module according to claim 7 or 8, wherein the imidazolium salts are salts of compounds defined by the formula (I):



wherein R<sub>1</sub> and R<sub>2</sub> independently denote a hydrogen atom or methyl; R<sub>3</sub> denotes methyl, ethyl, propyl, butyl, or hexyl.”

(26) All the description in claim 10 page 64 is amended as follows:

“The dye-sensitized solar cell module according to any one of claims 1 and 3 to 9, wherein either the first photoelectric conversion elements or the second photoelectric conversion elements contain

lithium iodide in the electrolytic layer thereof.”

(27) All the description in claim 11 on page 64 is amended as follows:

“The dye-sensitized solar cell module according to any one of claims 1 and 3 to 10, wherein the thicknesses of the porous photoelectric conversion layers of the first photoelectric conversion elements and the second photoelectric conversion elements differ.”

(28) In claim 12 on page 64, the phrase “claims 2 to 11” is amended to -- claims 1 and 3 to 11--.

(29) In claim 13 on page 64, the phrase “claims 2 to 12” is amended to -- claims 1 and 3 to 12--.

(30) All the description in claim 14 on page 65 is amended as follows:

“The dye-sensitized solar cell module according to any one of claims 1 and 3 to 13, wherein the light receiving surface areas of the respective porous photoelectric conversion layers of the first photoelectric conversion elements and the second photoelectric conversion elements differ.”

(31) All the description in claim 18 on page 65 is amended as follows:

“The dye-sensitized solar cell module according to any one of claims 1 and 3 to 17, wherein a first dye is adsorbed in the respective porous photoelectric conversion layers of a plurality of the first photoelectric conversion elements and a second dye different from the first dye is adsorbed in the respective porous photoelectric conversion layers of a plurality of the second photoelectric conversion elements.”

(32) In claim 19 on page 65, the phrase "claims 3 to 18" is amended to --claims 1, 4 and 18--.

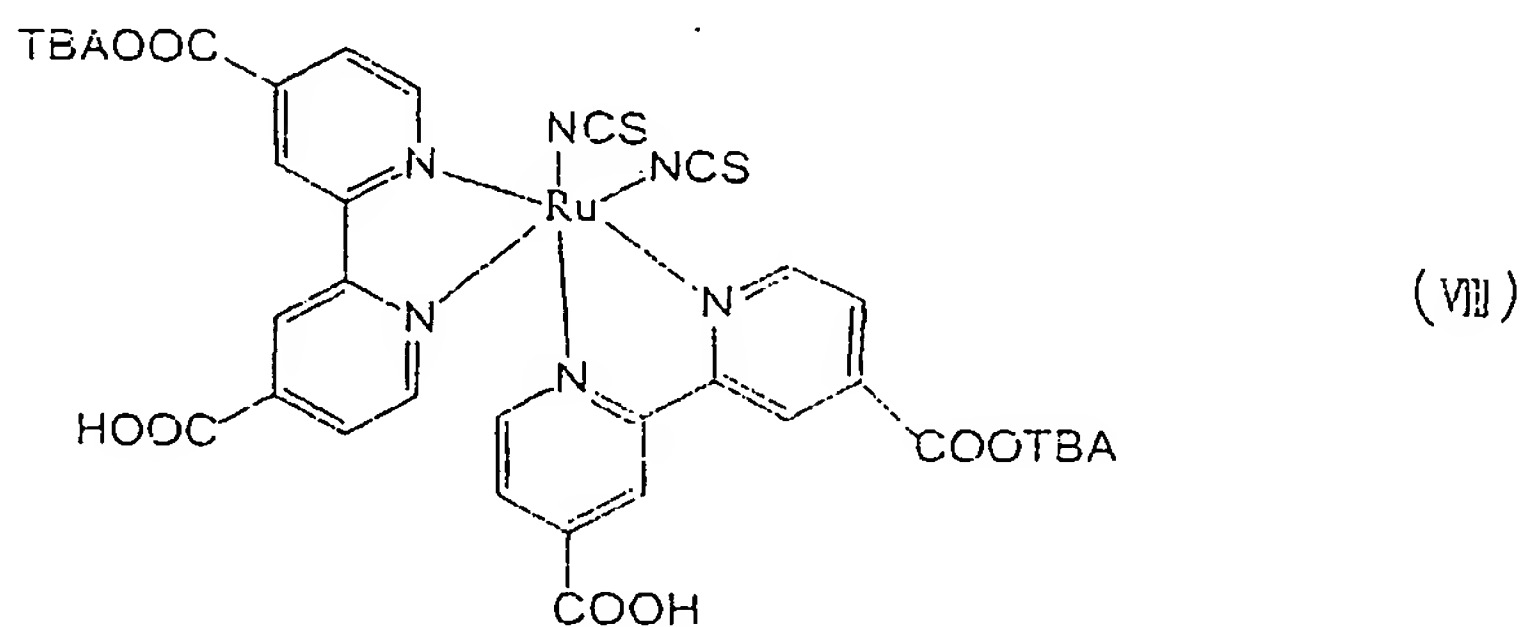
(33) In claim 21 on page 65, the phrase "claims 2 to 20" is amended to --claims 1 and 3 to 20--.

(34) In claim 25 on page 66, the phrase "claims 2 to 24" is amended to --claims 1 and 3 to 24--.

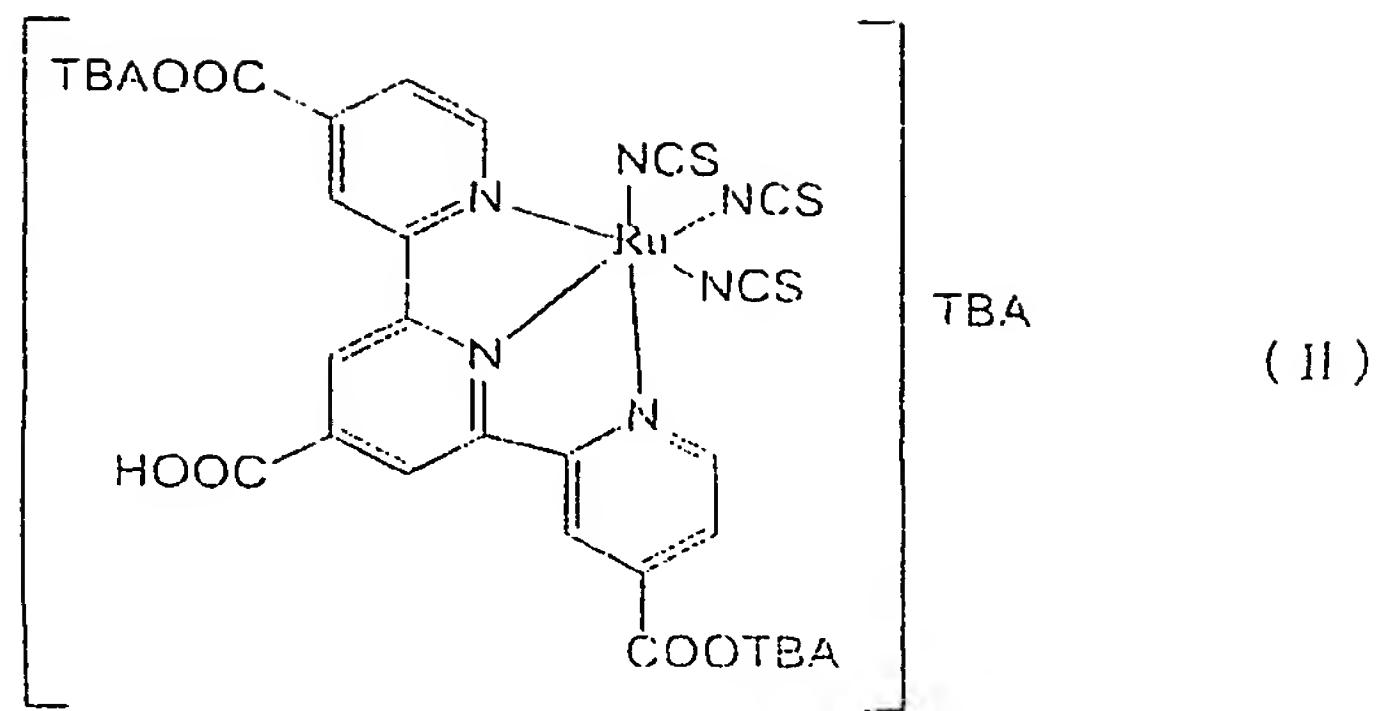
(35) In claim 30 on page 66, the phrase "claims 1 to 29" is amended to --claims 1 and 3 to 29--.

(36) As new claim 31, the following sentence is added under claim 30 on page 66, claim 31 stating:

"The dye-sensitized solar cell module according to claim 18, wherein a ruthenium dye defined by the formula (VIII):



is adsorbed on the respective porous photoelectric conversion layers of a plurality of the first photoelectric conversion elements and a black dye defined by the formula (II):



is adsorbed on the respective porous photoelectric conversion layers of a plurality of the second photoelectric conversion elements.”

solar cells composing the module cannot be exhibited at the maximum. In Fig. 8, the reference numerals 31 and 32 respectively denote a transparent substrate; 301, 302, and 303 respectively denote a transparent conductive film; 311, 312, and 313 denote an electrolytic solution; 321, 322, and 323 denote a porous titanium oxide layer; 331, 332, and 333 denote a catalyst layer; and 341, 342, and 343 denote an insulating layer.

## DISCLOSURE OF THE PRESENT INVENTION

### Problems to be Solved by the Invention

10           The present invention relates to a dye-sensitized solar cell module and provides a dye-sensitized solar cell with higher power than that of a conventional technique by improving constituent factors of a plurality of solar cells composing the module and accordingly solve the above-mentioned problems.

### 15   Means for Solving the Problems

          Accordingly, the present invention provides a dye-sensitized solar cell module comprising: first photoelectric conversion elements each comprising a transparent conductive layer, a porous photoelectric conversion layer adsorbing a dye, an electrolytic layer, a catalyst layer, and a conductive layer laminated in this order on a transparent substrate; second photoelectric conversion elements each comprising a transparent conductive layer, a catalyst layer, an electrolytic layer, a porous photoelectric conversion layer adsorbing a dye, and a conductive layer laminated in this order on the transparent substrate; and a supporting substrate formed on the respective conductive layers of the first and second photoelectric conversion elements, wherein one or more first photoelectric conversion elements and one or more second photoelectric conversion elements are alternately arranged in parallel between the transparent substrate and the supporting substrate, and the neighboring first photoelectric conversion elements and second photoelectric conversion elements are electrically connected in series, and wherein the first photoelectric conversion elements and the second

photoelectric conversion elements are different in at least one among the composition of the materials for the respective layers composing the photoelectric conversion elements; the particle diameters on the case that the materials of the respective layers are granular; the thickness and width of the respective layers; the form of the respective layers; and the dye.

#### Effects of the Invention

In the dye-sensitized solar cell module of the present invention, the first photoelectric conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the materials for the respective layers composing the photoelectric conversion elements; the particle diameters on the case that the materials of the respective layers are granular; the thickness and width of the respective layers; the form of the respective layers; and the dye, so that the properties such as the photoelectric conversion efficiency and the electric current can be improved as compared with a conventional technique.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a schematic cross-sectional view showing one row portion of an integrated dye-sensitized solar cell module of the present invention.

Fig. 2 is an explanatory drawing of a production method of the solar cell module shown in Fig. 1 and Fig. 2A is a schematic plain view showing the film formation state in the transparent insulating substrate X side and Fig. 2B is a schematic plain view showing the film formation state in the supporting substrate Y side.

Fig. 3 is a graph showing the correlation of the transmittance and absorbance of an electrolytic solution to be used in the dye-sensitized solar cell module of the present invention with the wavelength.

Fig. 4 is a graph showing the measurement results of the transmittance and absorbance of the electrolytic solution in

Embodiment 3 of the present invention.

Fig. 5 is a graph showing the correlation of ( $J_c/J_d$ ) with the module conversion efficiency in Embodiment 3 of the present invention.

Fig. 6 is a schematic cross-sectional view showing a  
5 dye-sensitized solar cell module of Embodiment 6 of the present invention.

Fig. 7 is a schematic cross-sectional view showing a conventional dye-sensitized solar cell.

Fig. 8 is a schematic cross-sectional view showing a  
10 conventional dye-sensitized solar cell.

#### BEST MODE FOR CARRYING OUT THE INVENTION

According to an embodiment of the present invention, there is provided a dye-sensitized solar cell (hereafter, sometimes referred to simply as a solar cell) comprising: first photoelectric conversion  
15 elements each comprising a transparent conductive layer, a porous photoelectric conversion layer adsorbing a dye, an electrolytic layer, a catalyst layer, and a conductive layer laminated in this order on a transparent substrate; second photoelectric conversion elements each  
20 comprising a transparent conductive layer, a catalyst layer, an electrolytic layer, a porous photoelectric conversion layer adsorbing a dye, and a conductive layer laminated in this order on the transparent substrate; and a supporting substrate formed on the respective  
conductive layers of the first and second photoelectric conversion elements, wherein one or more first photoelectric conversion elements  
25 and one or more second photoelectric conversion elements are alternately arranged in parallel between the transparent substrate and the supporting substrate, and the neighboring first photoelectric conversion elements and second photoelectric conversion elements are electrically connected in series, and wherein the first photoelectric  
30 conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the materials for the respective layers composing the photoelectric conversion elements;



the particle diameters on the case that the materials of the respective layers are granular; the thickness and width of the respective layers; the form of the respective layers; and the dye.

5 Next, the basic structure of the dye-sensitized solar cell module of the present invention will be described more practically with reference to Fig. 1. Fig. 1 is a schematic cross-sectional view showing one row portion of an integrated dye-sensitized solar cell module of the present invention.

The solar cell module (one row portion) M comprises a  
10 transparent insulating substrate X in the light receiving face side, a supporting substrate Y in the rear face (non-light receiving face) side, three first photoelectric conversion elements (a) installed between these substrates X and Y, two second photoelectric conversion elements (b) installed adjacent between the respective first photoelectric conversion  
15 elements

dye-sensitized solar cell is determined in accordance with the redox level (in the above-mentioned case  $I_3^-$  and  $I^-$ ) of the electrolytic solution and Fermi level of the porous photoelectric conversion layer (in the above-mentioned case, titanium oxide). Although the

5 above-mentioned electrolytic solution A and electrolytic solution B are different in the concentration of DMPII and the existence of LiI, generally lithium ion derived from LiI lowers the surface level of titanium oxide, so that the open circuit voltage value tends to be decreased in the case of the dye-sensitized solar cell. These

10 phenomena differ depending on the dye to be used and the porous photoelectric conversion layer and therefore it is made possible to produce a dye-sensitized solar cell module which generates higher output power than that of a conventional technique by adjusting the electrolytic solution compositions of the respective constituent

15 materials for the respective unit cells.

The ratio of the redox seeds such as  $I_3^-$  and  $I^-$  in the electrolytic solution is changed in accordance with the types of the imidazolium compounds such as DMPII.

Using EII, EMII, MPPII, HMII, and imidazole (hereinafter,

20 abbreviated as Imid. in some cases) as the imidazolium compounds other than DMPII, dye-sensitized solar cells are produced and the properties are evaluated. The electrolytic substance concentrations are adjusted as 0.6 mole/l for an imidazolium compound; 0.1 mole/l for LiI; 0.05 mole/l for  $I_2$ ; and 0.5 mole/l for TBP and acetonitrile is used as

25 a solvent.

The measurement is carried out while the light receiving face in the conductive layer side and the catalyst layer side of the respective dye-sensitized solar cells. The results in the case light receiving face is set in the conductive layer side are shown in Table 6 and the results in

30 the case light receiving face is set in the catalyst layer side are shown in Table 7.

[Table 6]

(Light receiving face: conductive layer side)

Imidazolium compound	EII	EMII	MPH	HMH
Short circuit current density (mA/cm <sup>2</sup> )	15.96	15.96	15.54	14.75
Open circuit voltage value (V)	0.627	0.655	0.671	0.674
FF	0.681	0.683	0.691	0.701
Conversion efficiency (%)	6.82	7.15	7.21	6.96
Imidazolium compound	Imid	DMPH (From Table 5)		
Short circuit current density (mA/cm <sup>2</sup> )	15.26	15.30		
Open circuit voltage value (V)	0.679	0.650		
FF	0.645	0.670		
Conversion efficiency (%)	6.68	6.66		

[Table 7]

(Light receiving face: catalyst layer side)

Imidazolium compound	EII	EMH	MPH	HMH
Short circuit current density (mA/cm <sup>2</sup> )	9.72	9.78	9.29	8.82
Open circuit voltage value (V)	0.611	0.642	0.654	0.653
FF	0.710	0.708	0.731	0.738
Conversion efficiency (%)	4.22	4.45	4.44	4.25
Imidazolium compound	Imid	DMPH (from Table 5)		
Short circuit current density (mA/cm <sup>2</sup> )	9.30	9.20		
Open circuit voltage value (V)	0.661	0.630		
FF	0.677	0.700		
Conversion efficiency (%)	4.16	4.06		

5 From Table 6 and Table 7, it is confirmed that the values of the respective properties are changed depending on the respective imidazolium compounds. Accordingly, it is made possible to produce

be extracted and the module conversion efficiency is considerably decreased more than the photoelectric conversion efficiency of a single dye-sensitized solar cell.

On the other hand, in a dye-sensitized solar cell, if the thickness  
5 of the photoelectric conversion layer is changed, the voltage and the current density is changed and if the thickness is made thick, the voltage tends to be low and the current density tends to increase. For example, in the case the thicknesses of photoelectric conversion layers are made the same, the short circuit current can efficiently be extracted  
10 and the open circuit voltage can be increased by making the thickness of single dye-sensitized solar cells in which the short circuit current density is increased thin and accordingly it is expected that the module conversion efficiency is increased.

In Embodiment 3, cell units are produced in the same manner as  
15 Embodiment 1, except that the electrolytic solution of the solar cell units and the thicknesses of the porous photoelectric conversion layers are made different.

As the electrolytic solution is used an electrolytic solution using acetonitrile as a solvent containing 0.6 mole/l of HMII, 0.1 mole/l for LiI,  
20 0.5 mole/l for TBP, and 0.01 mole/l for I<sub>2</sub>. On the other hand, the thicknesses of the porous photoelectric conversion layers are four types; 8, 12, 16, and 20  $\mu\text{m}$  and four types of dye-sensitized solar cells are produced. The measurement is carried out for the four types of the solar cells by radiating light to porous photoelectric conversion layer  
25 side and the catalyst layer side of the solar cells. The results in the case light receiving face is set in the porous photoelectric conversion layer side are shown in Table 10 and the results in the case light receiving face is set in the catalyst layer side are shown in Table 11.

setting a 30  $\mu\text{m}$ -thick separator between glass substrates with the same sizes and shapes of the actual dye-sensitized solar cell, injecting the electrolytic solution with the above-mentioned composition between the glass substrates, and radiating light to the glass substrates. As a  
5 result, with respect to the light rays with wavelength of about 300 to 550 nm, it is found that the transmittance generally tends to be decreased more as the wavelength becomes shorter. The decrease of the transmittance is supposed to considerably affect the short circuit current density in the case the light receiving face is set in the catalyst  
10 layer side.

In the case a dye-sensitized solar cell module with the structure as shown in Fig. 1 by using the above-mentioned unit cells, from the results of the experiments shown in Table 10 and Table 11, the short circuit current is about 29.0 mA when the light receiving face is set in  
15 the porous photoelectric conversion layer side and 23.2 mA when the light receiving face is set in the catalyst layer side if the thickness of the porous photoelectric conversion layer is 12  $\mu\text{m}$ . Therefore, in the case a series connection type dye-sensitized solar cell module is produced, the current of the module is controlled on the basis of the short circuit  
20 current value of the unit cells in which the light receiving face is set in the catalyst layer side and the short circuit current value of the unit cells in which the light receiving face is set in the porous photoelectric conversion layer side cannot be used advantageously and thus the conversion efficiency of the module is considerably decreased as  
25 compared with the photoelectric conversion efficiency of a cell unit.

Accordingly, based on the results of Table 10 and Table 11, while the thickness of the photoelectric conversion layer of the cell unit in which the light receiving face is set in the porous photoelectric conversion layer side is adjusted to be 12  $\mu\text{m}$  and the thickness of the  
30 photoelectric conversion layer of the cell unit in which the light receiving face is set in the catalyst layer side is adjusted to be 20  $\mu\text{m}$ , a dye-sensitized solar cell module comprising two cells connected in

series is produced. As a result, in the properties of the respective cell units themselves, the short circuit current density is 14.5 (mA/cm<sup>2</sup>) and the open circuit voltage value is 0.69 (V) for the unit cell in which the light receiving face is set in the porous photoelectric conversion layer side; the short circuit current density is 14.1 (mA/cm<sup>2</sup>) and the open circuit voltage value is 0.65 (V) for the unit cell in which the light receiving face is set in the catalyst layer side; and the short circuit current density is 14.4 (mA/cm<sup>2</sup>) and the open circuit voltage value is 1.33 (V) for the dye-sensitized solar cell module comprising two unit cells connected in series.

As described, the electric current values of respective unit cells are consequently advantageously utilized by making the thickness of the cell unit in which the light receiving face is set in the porous photoelectric conversion layer side different from the thickness of the cell unit in which the light receiving face is set in the catalyst layer side. Further, in the case the thickness of the photoelectric conversion layer of the cell unit in which the light receiving face is set in the porous photoelectric conversion layer side is adjusted to be 20 μm the same as that of the photoelectric conversion layer of the cell unit in which the light receiving face is set in the catalyst layer side, the open circuit voltage value is about 0.65 (V) according to the results shown in Table 10 and it is increased to 0.69 (V) by changing the thickness to be 12 μm. As a result, if the constitution of the dye-sensitized solar cell module of Embodiment 3 is employed, the ratio of the increase of the open circuit voltage is more significant than the ratio of the decrease of the photoelectric conversion efficiency due to the decrease of the current value of the module and accordingly the photoelectric conversion efficiency can be increased as a whole.

Further, the thickness of the photoelectric conversion layer of the cell unit in which the light receiving face is set in the porous photoelectric conversion layer side and the thickness of the photoelectric conversion layer of the cell unit in which the light receiving

face is set in the catalyst layer side are changed and the short circuit current density of the respective cell units and the photoelectric conversion efficiency of the module are measured. The relation between ( $J_c/J_d$ ) and  $\eta_m$  understood

5



Comparative Examples, however it is not intended that the present invention be limited to the illustrated Production Example, Examples, and Comparative Examples. In Examples and Comparative Examples, unless otherwise specified, dye-sensitized solar cells are produced  
5 under the conditions of Production Example.

(Example 15 and its Production Example)

A dye-sensitized solar cell module (reference to Fig. 1) in which five solar cell units were connected in series and integrated was produced in the same manner as Example 1, except that the electrolytic  
10 solutions for the electrolytic layers and the thickness of the porous photoelectric conversion layers were changed. Hereinafter, only the points of the production process of Example 15 different from Example 1 will be described below.

The thickness of the titanium oxide film as the porous  
15 photoelectric conversion layers was adjusted to be 12  $\mu\text{m}$  for the cell units in which the light receiving face was set in the porous photoelectric conversion layer side and to be 20  $\mu\text{m}$  for the cell units in which the light receiving face was set in the catalyst layer side.

As electrolytic solutions, acetonitrile was used as a solvent and  
20 an electrolytic solution was produced by dissolving 0.6 mole/l of HMII, 0.1 mole/l of LiI, 0.5 mole/l of TBP, and 0.01 mole/l of  $\text{I}_2$  in acetonitrile and the electrolytic solution was used.

The dye-sensitized solar cell module of Example 15 produced in the above-mentioned manner was put on a black stage in which the  
25 substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and the operation properties under the condition of simulating AM 1.5 sunlight were investigated. The results were 14.4 mA/cm<sup>2</sup> of the short circuit current density; 3.37 V of the open circuit voltage value; 0.66 of FF; and 6.41% of the module  
30 conversion efficiency.

(Examples 16)

The dye-sensitized solar cell module of Example 16 was

produced

scribed portions 21 of the substrates, the substrates X and Y subjected to the above-mentioned treatment were stuck to each other in the form shown in Fig. 1 and pressure bonded by heating at about 100°C for 10 minutes in an oven.

5           Successively, as an electrolytic solution to be used, an electrolytic solution was produced by dissolving 0.6 mole/l of DMPII (manufactured by Shikoku Chemicals Corp.), 0.1 mole/l of LiI (manufactured by Aldrich), 0.5 mole/l of TBP (manufactured by Aldrich), and 0.02 mole/l of I<sub>2</sub> (manufactured by Kishida Chemical Co., Ltd.) in  
10       acetonitrile as a solvent.

The electrolytic solution was injected into the unit cells of the dye-sensitized solar cell module shown in Fig. 1 by capillary effect and thereafter, the peripheral portions of the cells were sealed with an epoxy resin to produce the dye-sensitized solar cell module.

15           The dye-sensitized solar cell module of Example 17 produced in the above described manner was put on a black stage in which the substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and the operation properties under the condition of simulating AM 1.5 sunlight were investigated to find  
20       that the short circuit current density was 14.1 mA/cm<sup>2</sup>, the open circuit voltage value was 3.48 V, FF was 0.67, and the module conversion efficiency was 6.6%.

(Comparative Example 5)

25           A dye-sensitized solar cell module of Comparative Example 5 was produced in the same manner as Example 17, except that the width of the porous photoelectric conversion layers on both of the substrate X and the substrate Y in the series connection direction was adjusted to be 5.0 mm.

30           The dye-sensitized solar cell module of Comparative Example 5 produced in the above described manner was put on a black stage in which the substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and the operation

properties under the condition of simulating AM 1.5 sunlight were investigated to

The dye-sensitized solar cell module of Example 19 produced in the above described manner was put on a black stage in which the substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and the operation properties under the condition of simulating AM 1.5 sunlight were investigated to find that the short circuit current density was 11.9 mA/cm<sup>2</sup>, the open circuit voltage value was 3.38 V, FF was 0.67, and the module conversion efficiency was 5.4%.

(Comparative Example 6)

A dye-sensitized solar cell module of Comparative Example 6 was produced in the same manner as Example 17, except that the width of the porous photoelectric conversion layers on both of the substrate X and the substrate Y in the series connection direction was adjusted to be 5.0 mm.

The dye-sensitized solar cell module of Comparative Example 6 produced in the above described manner was put on a black stage in which the substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and the operation properties under the condition of simulating AM 1.5 sunlight were investigated to find that the short circuit current density was 12.0 mA/cm<sup>2</sup>, the open circuit voltage value was 3.38 V, FF was 0.62, and the module conversion efficiency was 5.0%.

(Example 20)

A dye-sensitized solar cell module of Example 20 was produced in the same manner as Example 17, except that the width of the porous photoelectric conversion layers on the substrate X in the series connection direction was adjusted to be 4.4 mm.

The dye-sensitized solar cell module of Example 20 produced in the above described manner was put on a black stage in which the substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and the operation properties under the condition of simulating AM 1.5 sunlight were investigated to find

that the

peripheral portions of the cells were sealed with an epoxy resin to produce the dye-sensitized solar cell module.

The produced dye-sensitized solar cell module was put on a black stage in which the substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and the operation properties under the condition of simulating AM 1.5 sunlight were investigated to find that the short circuit current density was 16.8 mA/cm<sup>2</sup>, the open circuit voltage value was 3.50 V, FF was 0.72, and the module conversion efficiency was 8.5%.

10 (Comparative Example 7)

A dye-sensitized solar cell module was produced in the same manner as the production example of Example 21, except that the black dye defined by the above-mentioned formula (II) was used for both of the supporting bodies X and Y. The produced dye-sensitized solar cell module was put on a black stage in which the substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and the operation properties under the condition of simulating AM 1.5 sunlight were investigated to find that the short circuit current density was 17.1 mA/cm<sup>2</sup>, the open circuit voltage value was 3.35 V, FF was 0.70, and the module conversion efficiency was 8.0%.

(Examples 22 to 24)

The dye-sensitized solar cell modules were produced in the same manner as Example 21, except that the dyes to be adsorbed were changed. The results are shown in Table 17. The dyes employed for the respective Examples are shown in Table 18.



[Table 17]

	Short circuit current density (mA/cm <sup>2</sup> )	Open circuit voltage value (V)	FF	Conversion efficiency (%)
Example 22	17.3	3.47	0.70	8.4
Example 23	11.6	3.11	0.68	4.9
Example 24	14.6	3.58	0.70	7.3

[Table 18]

	Supporting substrate X	Supporting substrate Y
Example 22	Dye VII	Dye II
Example 23	Dye III	Dye V
Example 24	Dye VI	Dye VII

5 (Example 25)

The solar cell module produced in Example 21 and the solar cell module produced in Comparative Example 7 were put on a black stage in which the substrate Y side was controlled to be at 25°C and the substrate X side was put in the light receiving face and a continuous irradiation experiment was carried out under the condition of simulating AM 1.5 sunlight radiation. The operation properties were measured after 150 hours to find that the conversion efficiency was 8.4% for the module of Example 21 and the conversion efficiency was 6.8% for the module of Comparative Example 7. It was confirmed that the amount of the electrolytic solution used in the module of Comparative Example 7 was decreased by eye observation. As described above, it is understood that integrated dye-sensitized solar cell modules of Examples 21 to 25 had the characteristics of the present invention and therefore, even in the case the size of the modules was as small as 59 mm × 65 mm, high power dye-sensitized solar cell modules

could be produced.

An electrolytic solution produced by dissolving 0.6 mole/l of DMPII, 0.1 mole/l of LiI, 0.05 mole/l of I<sub>2</sub>, and 0.5 mole/l of TBP in acetonitrile as a solvent was used as the electrolytic solution.

The current-voltage characteristic of the produced dye-sensitized solar cell module was investigated by setting the light receiving face in the substrate X under the condition of simulating AM 1.5 sunlight to find that the short circuit current density was 12.2 mA/cm<sup>2</sup>, the open circuit voltage value was 3.3 V, FF was 0.63, and the module conversion efficiency was 5.1%.

10 (Comparative Example 8)

A dye-sensitized solar cell module of Comparative Example 8 was produced in the same manner as Example 26, except that the titanium oxide paste containing scattered particles with an average particle diameter of 13 nm (trade name: T/SP, manufactured by Solaronix) was used for the semiconductor particles for the porous photoelectric conversion layers of all of the unit cells of the dye-sensitized solar cell module.

The current-voltage characteristic of the produced dye-sensitized solar cell module was investigated by setting the light receiving face in the substrate X under the condition of simulating AM 1.5 sunlight to find that the short circuit current density was 9.8 mA/cm<sup>2</sup>, the open circuit voltage value was 3.3 V, FF was 0.64, and the module conversion efficiency was 4.1%.

It is understood by comparison of Example 26 with Comparative Example 8, if the particle diameter of the semiconductor particles in the unit cells in which the light receiving face is set in the porous photoelectric conversion layer side and the particle diameter of the semiconductor particles in the unit cells in which the light receiving face is set in the catalyst layer side are made different, particularly if the diameter is made smaller in the porous photoelectric conversion layer side than in the catalyst layer side, a high conversion efficiency can be obtained.

5.4%.

It is understood from Example 27, if the porous photoelectric conversion layer is composed of a plurality of layers in the unit cells in which the light receiving face is set in the catalyst layer side and the  
5 respective layers are composed semiconductor particles with different particle diameters, a high conversion efficiency can be obtained.

(Examples 28 to 30)

Dye-sensitized solar cell modules of Examples 28 to 30 were produced in the same manner as Example 26, except the particle  
10 diameter of the semiconductor particles of the porous photoelectric conversion layers 2 in the light receiving face side was changed to be 20 nm (Example 28), 30 nm (Example 29), and 50 nm (Example 30).

Practically, in Example 28, a titanium oxide paste produced by dispersing 4.0 g of commercialized titanium oxide particles (trade name:  
15 Super Titania F5 grade, anatase type crystal; average particle diameter 20 nm; manufactured by Showa-Titanium Co., Ltd.) in 15 ml of diethylene glycol monomethyl ether by using glass beads and a paint shaker for 6 hours was used and the paste was applied by a doctor blade method, leveled at a room temperature for 1 hour, successively  
20 dried at 80°C in an oven, and fired at 500°C in air to form the porous semiconductor layer 2.

In Example 29, the porous semiconductor layer 2 was formed in the same manner as Example 28, except that titanium oxide particles (trade name: Super Titania F4 grade, anatase type crystal; average  
25 particle diameter 30 nm; manufactured by Tayca Corp.) were used.

In Example 30, the porous semiconductor layer 2 was formed in the same manner as Example 28, except that titanium oxide particles (trade name: Super Titania F5 grade, anatase type crystal; average particle diameter 50 nm; manufactured by Showa-Titanium Co., Ltd.)  
30 were used.

The current-voltage characteristic of the produced dye-sensitized solar cell modules of Examples 28 to 30 was investigated

by setting the

The operation properties of the produced dye-sensitized solar cell module were investigated under the condition of simulating AM 1.5 sunlight to find that the short circuit current density was 43.5 mA/cm<sup>2</sup>, the open circuit voltage value was 3.5 V, FF was 0.58, and the module  
5 conversion efficiency was 4.4%.

(Comparative Example 9)

The dye-sensitized solar cell module of Comparative Example 9 was produced in the same manner as Example 35, except catalyst layers 4 of the respective second photoelectric conversion elements (b) of  
10 the solar cell unit cells had no aperture part and the thickness was controlled to be 17 nm.

The operation properties of the produced dye-sensitized solar cell module were investigated under the condition of simulating AM 1.5 sunlight to find that the short circuit current density was 37.4 mA/cm<sup>2</sup>,  
15 the open circuit voltage value was 3.5 V, FF was 0.59, and the module conversion efficiency was 3.9%.

As described, even through having a module size as small as 55 mm × 65 mm, the integrated dye-sensitized solar cell modules of Examples 35 and 36 were found having high conversion efficiency since  
20 they had the characteristics of the present invention.

## CLAIMS

1. (AMENDED) A dye-sensitized solar cell module comprising:

5 first photoelectric conversion elements each comprising a transparent conductive layer, a porous photoelectric conversion layer adsorbing a dye, an electrolytic layer, a catalyst layer, and a conductive layer laminated in this order on a transparent substrate;

10 second photoelectric conversion elements each comprising a transparent conductive layer, a catalyst layer, an electrolytic layer, a porous photoelectric conversion layer adsorbing a dye, and a conductive layer laminated in this order on the transparent substrate; and

a supporting substrate formed on the respective conductive layers of the first and second photoelectric conversion elements,

15 wherein one or more first photoelectric conversion elements and one or more second photoelectric conversion elements are alternately arranged in parallel between the transparent substrate and the supporting substrate, and the neighboring first photoelectric conversion elements and second photoelectric conversion elements are electrically connected in series, and

20 wherein the first photoelectric conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the materials for the respective layers composing the photoelectric conversion elements; the particle diameters on the case that the materials of the respective layers are granular; the thickness and width of the respective layers; the form of  
25 the respective layers; and the dye.

2. Cancelled

30 3. (AMENDED) The dye-sensitized solar cell module according to claim 1, wherein a short circuit current of the second photoelectric conversion elements in the case where a light receiving



face thereof is set in the porous photoelectric conversion layer side opposite the catalyst layer side is greater than a short circuit current of the first photoelectric conversion elements in the case where a light receiving face thereof is set in the porous photoelectric conversion layer side opposite the catalyst layer side.

4. (AMENDED) The dye-sensitized solar cell module according to claim 1 or 3, wherein the first photoelectric conversion elements and the second photoelectric conversion elements are different in at least one among the composition of the electrolytic layers; the thickness of the porous photoelectric conversion layers; the width of the porous photoelectric conversion layers; the average particle diameter of the semiconductor particles composing the porous photoelectric conversion layers; and the form of the catalyst layers.

5. The dye-sensitized solar cell module according to claim 4, wherein the first photoelectric conversion elements and the second photoelectric conversion elements contain iodine in the respective electrolytic layers and the iodine concentration in the electrolytic layers of the second photoelectric conversion elements is lower than the iodine concentration in the electrolytic layers of the first photoelectric conversion elements.

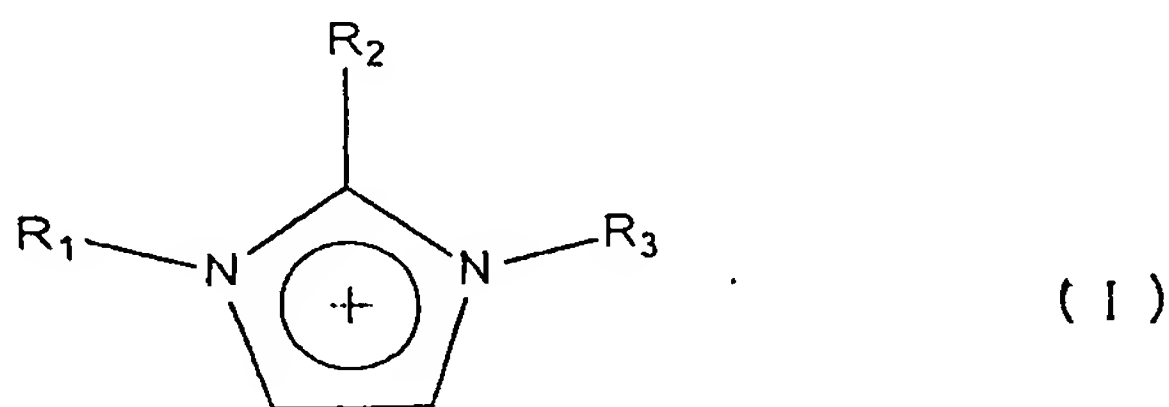
6. The dye-sensitized solar cell module according to claim 5, wherein the ratio  $M1/M2$  of the iodine concentration  $M1$  in the electrolytic layers of the first photoelectric conversion elements and the iodine concentration  $M2$  in the electrolytic layers of the second photoelectric conversion elements is higher than 1 and not higher than 5.

7. (AMENDED) The dye-sensitized solar cell module according to any one of claims 1 and 3 to 6, wherein the first photoelectric conversion elements and the second photoelectric

conversion elements respectively contain imidazoles or imidazolium salts in their electrolytic layers, the imidazole or imidazolium salt contained in each of the electrolytic layers of the first photoelectric conversion elements being different from that contained in each of the electrolytic layers of the second photoelectric conversion elements.

8. (AMENDED) The dye-sensitized solar cell module according to any one of claims 1 and 3 to 7, wherein the imidazoles or imidazolium salts contained in the respective electrolytic layers of the first photoelectric conversion elements and the second photoelectric conversion elements differ in concentration.

9. (AMENDED) The dye-sensitized solar cell module according to claim 7 or 8, wherein the imidazolium salts are salts of compounds defined by the formula (I):



wherein R<sub>1</sub> and R<sub>2</sub> independently denote a hydrogen atom or methyl; R<sub>3</sub> denotes methyl, ethyl, propyl, butyl, or hexyl.

20

10. (AMENDED) The dye-sensitized solar cell module according to any one of claims 1 and 3 to 9, wherein either the first photoelectric conversion elements or the second photoelectric conversion elements contain lithium iodide in the electrolytic layer thereof.

25

11. (AMENDED) The dye-sensitized solar cell module according to any one of claims 1 and 3 to 10, wherein the thicknesses of

the porous photoelectric conversion layers of the first photoelectric conversion elements and the second photoelectric conversion elements differ.

5           12. (AMENDED)       The dye-sensitized solar cell module  
according to any one of claims 1 and 3 to 11, wherein the thicknesses of  
the porous photoelectric conversion layers of the first photoelectric  
conversion elements are thinner than the thicknesses of the porous  
photoelectric conversion layers of the second photoelectric conversion  
10 elements.

          13. (AMENDED)       The dye-sensitized solar cell module  
according to any one of claims 1 and 3 to 12, wherein when the short  
circuit current density of the first photoelectric conversion elements is  
15 defined as  $J_d$  and the short circuit current density of the second  
photoelectric conversion elements is defined as  $J_c$ ,  $(J_c/J_d) > 0.7$  is  
satisfied.

          14. (AMENDED)       The dye-sensitized solar cell module  
20 according to any one of claims 1 and 3 to 13, wherein the light receiving  
surface areas of the respective porous photoelectric conversion layers of  
the first photoelectric conversion elements and the second photoelectric  
conversion elements differ.

25           15.       The dye-sensitized solar cell module according to claim  
14, wherein the light receiving surface areas of the respective porous  
photoelectric conversion layers of the second photoelectric conversion  
elements are larger than the light receiving surface areas of the  
respective porous photoelectric conversion layers of the first  
30 photoelectric conversion elements.

          16.       The dye-sensitized solar cell module according to 14 or

15, wherein the widths of the respective porous photoelectric conversion layers of the first photoelectric conversion elements and the second photoelectric conversion elements differ in the series connection direction of the solar cells.

5

17. The dye-sensitized solar cell module according to any one of claims 14 to 16, wherein the light receiving surface areas of the respective porous photoelectric conversion layers of a plurality of the first photoelectric conversion elements are the same and the light  
10 receiving surface areas of the respective porous photoelectric conversion layers of a plurality of the second photoelectric conversion elements are the same.

18. (AMENDED) The dye-sensitized solar cell module  
15 according to any one of claims 1 and 3 to 17, wherein a first dye is adsorbed in the respective porous photoelectric conversion layers of a plurality of the first photoelectric conversion elements and a second dye different from the first dye is adsorbed in the respective porous photoelectric conversion layers of a plurality of the second photoelectric  
20 conversion elements.

19. (AMENDED) The dye-sensitized solar cell module  
according to any one of claims 1, 4 and 18, wherein an open circuit voltage of the first photoelectric conversion elements and an open  
25 circuit voltage of the second photoelectric conversion elements differ.

20. The dye-sensitized solar cell module according to claim 19, wherein open circuit voltage values of the first photoelectric conversion elements are higher than open circuit voltage values of the  
30 second photoelectric conversion elements.

21. (AMENDED) The dye-sensitized solar cell module

according to any one of claims 1 and 3 to 20, wherein the average particle diameter of the semiconductor particles of the porous semiconductor layers of the first photoelectric conversion elements is smaller than the average particle diameter of the semiconductor particles of the porous semiconductor layers of the second photoelectric conversion elements.

22. The dye-sensitized solar cell module according to claim 21, wherein the porous semiconductor layer of at least each of the second photoelectric conversion elements is composed of a plurality of layers and the average particle diameter of the semiconductor particles in the porous semiconductor layer closest to the supporting substrate is larger than the average particle diameter of the semiconductor particles in the porous semiconductor layer farthest from the supporting substrate.

23. The dye-sensitized solar cell module according to claim 22, wherein the average particle diameter of the semiconductor particles of the porous semiconductor layers of the first photoelectric conversion elements is 30 nm or smaller and the semiconductor particles with a particle diameter of 100 nm or larger are contained in the porous semiconductor layers of the second photoelectric conversion elements.

24. The dye-sensitized solar cell module according to claim 23, wherein the porous semiconductor layer of each of the second photoelectric conversion elements is composed of a plurality of layers and the semiconductor particles with a particle diameter of 100 nm or larger are contained in the porous semiconductor layer closest to the supporting substrate and the semiconductor particles with an average particle diameter of 30 nm or smaller are contained in the porous semiconductor layer farthest from the supporting substrate.

25. (AMENDED) The dye-sensitized solar cell module according to any one of claims 1 and 3 to 24, wherein the light transmittance of the catalyst layers of the second photoelectric conversion elements is lower than the light transmittance of the catalyst layers of the first photoelectric conversion elements since the shapes of the catalyst layers of the first photoelectric conversion elements and the catalyst layers of the second photoelectric conversion elements differ.

10 26. The dye-sensitized solar cell module according to claim 25, wherein the catalyst layers of the second photoelectric conversion elements have aperture parts.

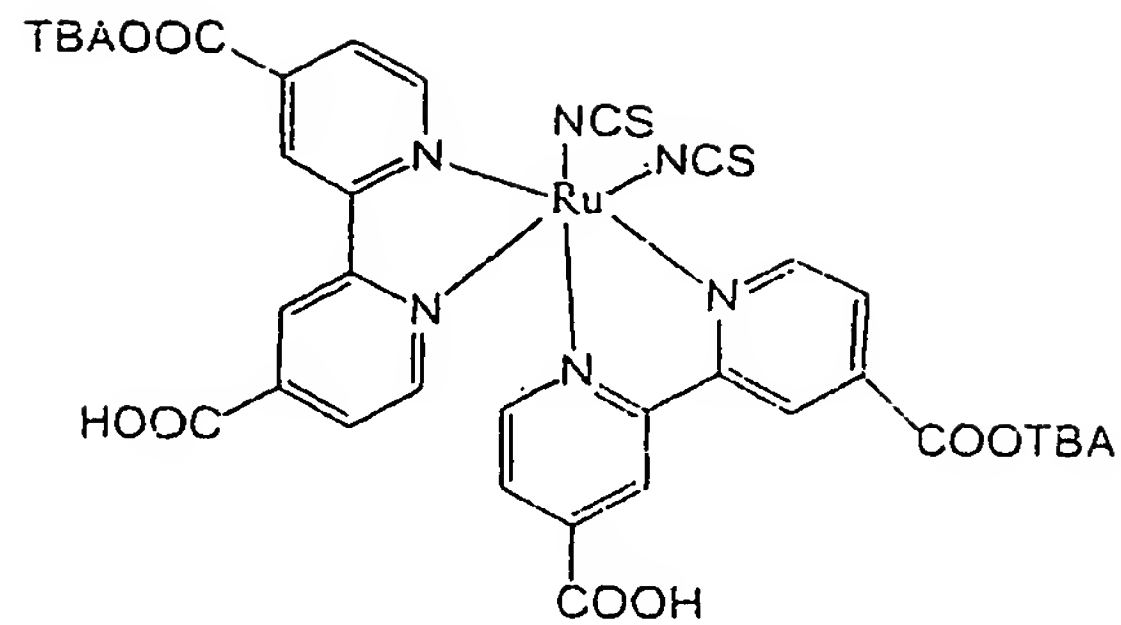
15 27. The dye-sensitized solar cell module according to claim 26, wherein the catalyst layers of the second photoelectric conversion elements have a lattice-like shape.

20 28. The dye-sensitized solar cell module according to claim 26, wherein the catalyst layers of the second photoelectric conversion elements have a stripe shape.

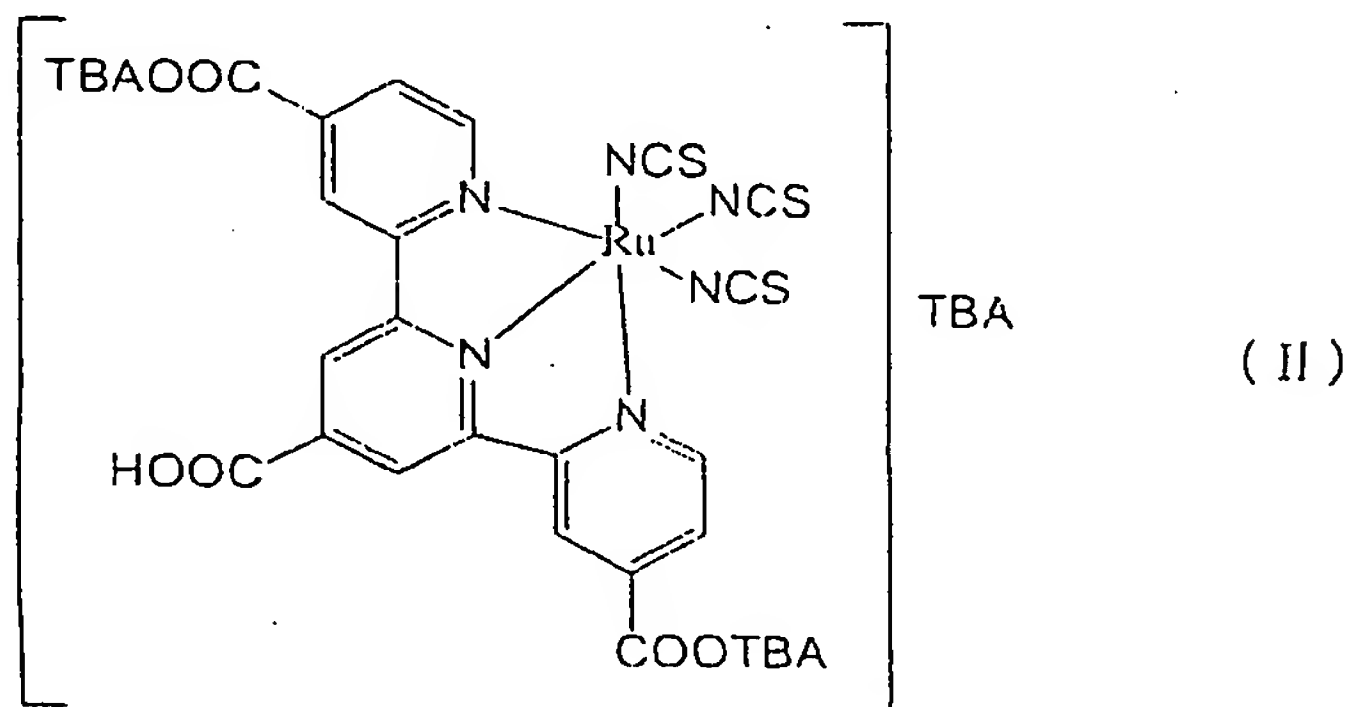
25 29. The dye-sensitized solar cell module according to claim 26, wherein the catalyst layers of the second photoelectric conversion elements have a dotted shape.

30. (AMENDED) The dye-sensitized solar cell module according to any one of claims 1 and 3 to 29, wherein the catalyst layers contain Pt.

30 31. (NEW) The dye-sensitized solar cell module according to claim 18, wherein a ruthenium dye defined by the formula (VIII):



is adsorbed on the respective porous photoelectric conversion layers of a plurality of the first photoelectric conversion elements and a black dye defined by the formula (II):



5

is adsorbed on the respective porous photoelectric conversion layers of a plurality of the second photoelectric conversion elements.